Assessing the effect of broadleaf woodland expansion on acidic dry deposition and streamwater acidification

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Abstract

The study aim was to determine whether enhanced dry deposition of acidic atmospheric pollutants by broadleaf woodland expansion could increase the potential for acidification of surface waters in acid-sensitive areas. Dry sulphur (S) and nitrogen (N) deposition was modelled with the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model using a roughness length value calculated specifically for birchwoods. Two scenarios were investigated for an acid-sensitive area in Scotland where broadleaf woodland expansion, mainly as birchwood, is occurring: (1) 2002 emissions and broadleaf woodland cover of 5.6%; (2) 2020 projected emissions and broadleaf cover of 29%. The roughness length calculated for birch with Raupach's simplified drag-partition model was 0.73 m, lower than the value of 1.0 m for conifers which is the default for forest land cover in FRAME. Modelled dry S and N deposition increased between 2002 and 2020 from 8.7 to 29 x 10^{-3} keq ha^{-1} $year^{-1}$ of H^+. However, modelled total dry and wet non-marine S and N deposition decreased during the same period from 1070 to 682 x 10^{-3} keq ha^{-1} $year^{-1}$ of H^+ due to the lower projected emissions in 2020 and the dominance of wet deposition in the remote and upland study area (mean annual rainfall 2275 mm). The modelled total non-marine S and N deposition was used to calculate streamwater critical loads exceedance with the First-order Acidity Balance (FAB) model for five catchments in the study area. The modelled deposition for both the 2002 and 2020 scenarios was less than the calculated streamwater critical loads so the catchments were not considered at risk of streamwater acidification under the projected future emissions and increased broadleaf woodland cover. Nevertheless, broadleaf expansion could pose a greater risk of acidification in acid-sensitive areas with lower rainfall, closer to pollutant sources, where dry deposition accounts for a higher proportion of total S and N deposition.

Keywords broadleaf woodland expansion; critical loads; dry deposition; Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model; First-order Acidity Balance (FAB) model; roughness length.

1. Introduction

Freshwaters continue to suffer from acidification in many parts of the world (e.g., Kowalik et al., 2007; Lawrence et al., 2008; Nakahara et al., 2010) due to a combination of present-day or historic high acid deposition and the occurrence of soils with low buffering capacity, resulting in adverse impacts on the aquatic ecology (Baldigo et al., 2009; Harriman et al., 1987). Surface water acidification is predominantly caused by the transfer of anthropogenically-derived strong acids and acid-forming substances, in particular sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and the particulate acidifying compounds sulphate (SO_4) and nitrate (NO_3), from the atmosphere via deposition to the earth's surface (Driscoll et al., 2001). Pollutants are deposited in rainwater and snow (wet deposition), cloud water (occult deposition) and directly onto plant and other surfaces (dry deposition). Acid deposition in the UK consists mainly of wet deposition of SO_4, NO_3 and ammonium (NH_4) and dry deposition of the gases, SO_2, nitrogen dioxide (NO_2), nitric acid (HNO_3) and NH_3, and aerosols containing SO_4, NO_3 and NH_4 (RoTAP, in press). In soils with low buffering capacity, incoming acid anions can increase water acidity and lead, through ion exchange, to the leaching from soils to freshwaters of cations (e.g., Al^{3+}) which can be toxic to fish and other biota. In areas of high acid deposition, forests can enhance the dry and occult deposition of S and N (termed the scavenging effect) due to the greater turbulence caused by the stand structure as compared to low vegetation (Fowler et al., 1989), and subsequently exacerbate acidification in sensitive surface waters. The scavenging effect increases with tree height, and therefore forest age, and with altitude (Nisbet et al., 1995).

Reducing emissions of acidic pollutants is the most effective way of solving the problem of acidification and internationally this is being addressed through the Convention on Long-Range Transboundary Air Pollution (CLRTAP), established in 1979 under the auspices of the United Nations Economic Commission for Europe (UNECE), and its associated Protocol to Abate Eutrophication, Acidification and Ground-Level Ozone, which was agreed in 1999. As a result, S-SO_2 and N-NO_2 emissions in the UK decreased from 1970 to 2005 by 89% and 48%, respectively, with deposition of total (wet and dry) oxidised sulphur (S-SO_x) and the sum of NO_x and reduced forms of nitrogen (NH_y) declining by 62% and 12%, respectively, between 1990 and 2005 (Matejko et al., 2009). Streamwaters within acid-sensitive areas are displaying clear signs of chemical recovery, although the biological response has been relatively muted to date (Kernan et al., 2010). Whilst further reductions in S and N deposition are likely, forest expansion may result in increased scavenging and deposition of the remaining atmospheric acidic pollutants. To identify areas at potential risk of acidification as a result of forest expansion the critical loads approach was used in the current study. The approach has been applied to assess the risk of freshwater acidification in countries that have ratified the CLRTAP and has been incorporated into the Forests & Water Guidelines (Forestry Commission, 2003) which describe best practice for minimising the effect of forestry activities on the freshwater environment in the UK. The critical load of acidity for surface waters is “the highest deposition of acidifying compounds that will not cause chemical changes leading to long term harmful effects on ecosystem structure and function” (Nilsson and Grennfelt, 1988). The critical load is widely calculated with steady-state models (described later) which consider the pre-acidification availability of base cations, estimated from present-day water chemistry, and the charge balance of S and N in catchments, minus a required level of buffering or acid neutralising capacity (ANC) to maintain suitable conditions. An appropriate ANC threshold is selected to maintain acceptable conditions for specified aquatic organisms (usually fish). Surface waters that receive acid deposition greater than the critical load are termed “exceeded” and at risk of biological damage. Acid deposition inputs for critical load exceedance calculations are typically obtained from national- or continental-scale atmospheric transport and chemistry models which simulate the emission, diffusion and advection of pollutant gases, their chemical transformation in the atmosphere, washout by precipitation and dry deposition on to vegetation (Dore et al., 2007).

In the UK conifer afforestation has declined since the late 1980s (Farmer and Nisbet, 2004) and the majority of new forest planting is broadleaf. In Scotland the aim is to increase woodland cover from 17 to 25% by the second half of this century which would involve creating 650,000 ha of new woodland (Forestry Commission Scotland, 2009), much of which is likely to be broadleaf. The scavenging effect is expected to be less for broadleaf woodland than for the more aerodynamically rough conifer canopies, but the impact of broadleaf expansion over large areas could delay the recovery of acidified surface waters, or even lead to further acidification in the most sensitive areas (Alexander and Cresser, 1995).

This study assessed the risk of streamwater acidification arising from enhanced dry acid deposition due to planned broadleaf woodland expansion, focussing on birch canopies in an acid-sensitive area in central Scotland. Two scenarios were investigated: a 2002 scenario using the early 2000s broadleaf cover and acid deposition generated from 2002 emissions data, and a future scenario using projected emissions data for 2020 and the higher level of 29% broadleaf woodland cover planned for 2020. In both scenarios dry deposition onto birch was modelled using a value of the roughness length calculated specifically for birch, which was then combined with the modelled wet deposition to assess the risk of streamwater acidification using the critical loads methodology.

2. Methodology

2.1. Study catchments

Five catchments (LK1, LK2, LK3, LK4 and LK5) close to the north-western shore of Loch Katrine, at the eastern edge of the Loch Lomond and Trossachs National Park, Scotland (Fig. 1) were selected to assess the risk to streamwater acidification by acid deposition under 2002 and 2020 emissions and woodland cover scenarios. Freshwaters in this area drain base-poor soils and rock and are classified as potentially at risk of a forestry acidification effect according to the critical loads exceedance map for UK freshwaters used in the Forests & Water Guidelines (Forestry Commission, 2003). Consequently, a site-specific critical load assessment is required if more than 10% of any stream catchment is planted or restocked with conifers or more than 30% with broadleaves (Forestry Commission, 2003). The characteristics of the catchments are detailed in Gagkas (2007) (and summarised in Table 1). All catchments have an upland character and are underlain by a geology comprising Dalradian schists, grits and shales (BGS, 1995) and covered predominantly by ortsteinic albic folic podzols, histic podzols and histic leptosols (nomenclature after IUSS Working Group WRB, 2006). Mean annual rainfall (1961-2005) was 2275 mm (British Atmospheric Data Centre, BADC). Land cover in the study area was estimated from digitised aerial imagery taken in 2000 (1: 10 000 scale, Forest Enterprise, Aberfoyle, Scotland) and considered representative of conditions in 2002. Woodland cover, estimated by digitising woodland patches and individual trees in ArcGIS (ESRI, CA, USA), ranged between 2.64 and 20.6% in the study catchments, with the remaining vegetation cover comprising acid grassland and blanket bog. Most of the woodland in the catchments occurred at 250-350 m elevation and was dominated by an even-aged stand of downy birch (*Betula pubescens*) with a very open canopy. Forestry Commission Scotland manages the land around Loch Katrine and aims to increase the native woodland cover from 890 ha to 2890 ha by 2028, of which an estimated 1150 ha will be planted (58% of the area of expansion), with the remainder achieved through natural regeneration. Birch was selected as the tree species of focus for this study since

Betula spp. are one of the more important broadleaf genera in new native woodland planting and natural regeneration schemes in Scotland and will cover 635 ha of the 2000 ha native woodland expansion scheme in the Loch Katrine area (Forestry Commission Scotland, 2008).

Figure 1, Table 1

2.2. Atmospheric acid deposition modelling

Wet deposition of acidic atmospheric species is unaffected by land cover, unlike dry and occult deposition. Wet and dry deposition of oxidised S ($SO_x = SO_2 + SO_4^{2-}$), oxidised N ($NO_y = NO + NO_2 + NO_3^- + HNO_3$ + PAN (peroxyacetyl nitrate)) and reduced N ($NH_x = NH_3 + NH_4^+$) to a 5 km x 5 km grid square including the study catchments were calculated individually using the Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model (Singles et al., 1998; Vieno et al., 2010). FRAME is a statistical Lagrangian trajectory model which is used in government policy applications in the UK to estimate the effects of pollutant gas abatement on the exceedance of critical loads for acidification and eutrophication (e.g., Dore et al., 2007). Woodland cover in the grid square was determined as described in 2.1 since this provided a more accurate representation than using existing woodland polygons at a lower spatial resolution. Downy birch was the dominant woodland species, covering 139 ha or 5.6% of the grid square. Other tree species (conifer and mixed broadleaves) occupied a further 0.4 ha, with the remainder of the grid square covered mostly by grassland and bare rock (2291 ha or 92% cover) or open water (Loch Katrine). The planned native woodland expansion will increase the woodland cover in the grid square as a whole to 725 ha or 29%. It was assumed that the woodland would replace grassland and comprise mainly birch, as the local dominant pioneer species, and be fully established by 2020 (Fig. 1). In the individual study catchments the projected areas of broadleaf woodland in 2020 will exceed 30% of catchment area (34-58% of catchment area, apart from

LK4 at 24% of catchment area), the threshold above which catchment-specific critical loads assessments are required.

Deposition of SO_2, NO_y and NH_3 to the grid square was calculated in FRAME from the estimated emissions compiled in the UK National Atmospheric Emissions Inventory (www.naei.org.uk). Wet deposition of chemical species was calculated in FRAME using average annual precipitation and a scavenging coefficient, to represent the amount removed from the air column by rain (but not by vegetation), and also incorporated a seeder-feeder process (Fowler et al., 1988; Dore et al., 2006a) to account for orographic enhancement of wet deposition (Fournier et al., 2005). Although occult deposition is not explicitly modelled, it is significant only for forests at moderate–high altitudes (>600 m) in the western UK which are frequently enveloped in cloud (Fowler et al., 1989; RoTAP, in press). Dry deposition of SO_2, NO_y and NH_3 are calculated in FRAME individually for five different land cover types (arable, forest, moorland, grassland, and urban) using a canopy resistance model (Smith et al., 2000). Dry deposition is first generated assuming 100% cover of each land cover type and then a spatially-weighted average is calculated based on the actual land cover mix within the grid square. The model assumes that the deposition velocity (V_d) of pollutants is the inverse of the sum of three resistances in series: the aerodynamic resistance (R_a), the laminar boundary layer resistance (R_b), and the surface resistance (R_c):

$$V_d = \left(R_a + R_b + R_c\right)^{-1} \qquad \text{Eq. (1)}$$

The roughness length of the canopy, z_0, used with wind speed to calculate R_a and R_b (Smith et al., 2000), is the only vegetation-specific parameter in FRAME that can be altered and was modified to reflect the effect of birch woodland expansion upon dry deposition to the study grid square. The default z_0 value for forest in FRAME, 1.0 m, is for conifer forest (Singles, 1996) and was considered too high for broadleaf woodland due to the smaller leaf surface area and shedding of leaves in winter (Robertson et al., 2000). Instead, a value of z_0 specific to birch woodland in the study catchments was calculated (see the Appendix) using Raupach's drag-partition model (R94 version, Raupach, 1994), as in the ForestGALES model (Gardiner et al., 2000), and the mean (n=50) measured birch tree height in the study catchments of 10.2 m. Since the calculation of R_c does not involve any parameters related to canopy roughness, in this study the default FRAME parameterisation for forest was used, apart from the value for NH_3 of 20 s m^{-1} (Singles et al., 1998) which was replaced by 80 s m^{-1}, the R_c value for deciduous trees used in the UK Met Office Rainfall and Evaporation Calculation System (MORECS, Hough and Jones, 1997).

2.3. Assessment of streamwater acid-sensitivity using the critical loads methodology

2.3.1. Streamwater sampling and chemical analysis

Critical loads in the study catchments were assessed following the protocol of the Forestry Commission (2003), in which streamwater was sampled during high flow conditions when streamwater is expected to be most acidic. Samples were taken at the outlet of each catchment in acid-washed polyethylene bottles on a total of 9-10 occasions in the 6 months comprising February, March, April, October and November 2005 and February 2006. pH was measured on site (Hanna Instruments 9025 pH meter) and samples were stored in the dark at 4 °C prior to analysis. Gran alkalinity was determined within 24 hours of sampling by manual titration with 0.01 M HCl from pH 4.5 to 3.5 (Neal, 2001). Base cations (Ca, Mg, Na and K) were determined by flame atomic absorption spectrometry (Unicam AA M Series), Cl and SO_4 by liquid ion chromatography (Dionex DX-500) and NO_3 by continuous flow analysis (Bran & Luebbe AA3). Dissolved organic carbon (DOC) was determined in filtered (0.45 μm) samples from one occasion (in November 2005) using a Thermalox Analyser. Standard laboratory quality assurance measures detailed in Gagkas (2007) provided confidence in the accuracy, precision and reproducibility of the streamwater analyses.

2.3.2. Calculation of critical load exceedance

Critical load exceedance (*Ex(FAB)*) for each catchment for 2002 and 2020 deposition scenarios was calculated from the streamwater chemistry results using the First-order Acidity Balance (FAB) model (Posch et al., 1997), a steady-state charge balance approach that is used in the UK to calculate freshwater critical loads to guide emissions policy (UK National Focal Centre, 2004). The strength of the FAB model compared to other steady state models is that it contains a more detailed formulation of the long-term sustainable sinks for deposited N. The formulation of the FAB model used in this study was:

$$Ex(FAB) = \left(S_{dep} + N_{dep}\right) - \left(N_{imm} + N_{den}\right) - AA_{leach} \qquad \text{Eq. (2)}$$

where all units are in equivalents per unit area and time and:

S_{dep} = non-marine total S deposition

N_{dep} = total N (oxidised and reduced) deposition

N_{imm} = long term immobilisation of N in the terrestrial catchment, taking account of fixation and organic N export from the catchment

N_{den} = N lost through denitrification in catchment soils

AA_{leach} = acid anion leaching from the catchment

The FAB model terms for the removal offsite of N in harvested forest vegetation and retention of N and S in lakes were omitted from Eq. (2) since the native woodland would not be felled and the study catchments did not contain lakes. The assumptions underlying Eq. (2) are that: long-term sinks of S in catchment soils and vegetation are negligible; there are no significant N inputs other than atmospheric deposition; and NH_4^+ leaching is negligible because any inputs are taken up by biota, adsorbed by soil, or converted to NO_3 (Posch et al., 1997). Estimates of S_{dep} and N_{dep} were obtained from the FRAME model output; N_{imm} and N_{den} were estimated by multiplying default values for specified soil types (Hall et al., 1998) by the relative proportion of the catchment area underlain by each soil type (Table 1). Due to similar soil type composition, the mean N_{imm} and N_{den} values varied little between the study catchments at 0.19-0.21 and 0.07-0.09 keq ha^{-1} year^{-1} of H^+, respectively. The FAB model term AA_{leach} is equivalent to the critical load (CL) calculated with the Steady-State Water Chemistry (SSWC) model (Henriksen et al., 1986) (Eq. 3):

$$CL = \left([BC]_0^* - [ANC]_{crit}\right) \times Q \qquad \text{Eq. (3)}$$

The CL of acidity (expressed as an annual flux, keq ha^{-1} year^{-1} of H^+) is based on the principle that the acid load to water should not exceed the long-term supply of neutralising base cations in the catchment derived from weathering (represented by ($[BC]_0^*$) minus a critical acid neutralising capacity (ANC_{crit}) set to protect selected biota (Henriksen et al., 1986). This study used a value of ANC_{crit} = 0 μeq l^{-1}, which provides a 50% probability of protecting brown trout (*Salmo trutta*) populations when applied to mean streamwater chemistry conditions (UK National Focal Centre, 2004), but 90% or greater protection for high flows .Whilst the chosen ANC_{crit} value is lower than that applied elsewhere (e.g. a value of 20 μeq l^{-1} is adopted by the UK National Focal Centre (2003)), it is considered appropriate for critical load exceedance calculations based on high flow water samples, as recommended by the Forests & Water Guidelines, the UK forestry guidance. The CL for each catchment was calculated in the FAB model using the mean high flow streamwater chemistry. Non-marine solute concentrations were estimated using published seasalt correction factors based on the ratio of ions to chloride (UBA, 2004). Streamwater ANC was determined in μeq l^{-1} as the difference between the measured streamwater base cation (Ca, Mg, Na, K) and acid anion (Cl, SO_4, NO_3) concentrations. Concentrations were converted to fluxes using catchment runoff (Q), estimated as 85% of the annual rainfall (2005, the main streamwater sampling period), as recommended by the UK Critical Loads Advisory Group and adopted by the Forests & Water Guidelines (Forestry Commission, 2003) for catchments where Q has not been measured. The ANC_{crit} value chosen and the runoff estimation method used in this study are subject to debate (e.g. Lydersen et al. (2004) indicate that the ANC_{crit} value selected should take account of the organic carbon concentration). However, in a sensitivity analysis of CLs calculated using the FAB model, Gagkas et al. (2010) showed that, for 14 acid-sensitive UK upland catchments, the CL values were relatively insensitive to different runoff estimates and different ANC_{crit} values, being insufficient to shift the status of any of the catchments from protected to exceeded.

3. Results

3.1. Modelled deposition

The roughness length for the birchwoods, z_{0bir}, 0.73 m, calculated using the R94 version of Raupach's drag-partition model (Raupach, 1994) was, as expected, smaller than the default value of 1.0 m for conifer forest used in FRAME. The z_{0bir} value was used to calculate R_a and R_b in the canopy resistance model to yield dry deposition estimates of SO_x, NO_y and NH_x. Modelled SO_x and NH_x deposition for 2002 with z_{0bir} was consequently lower than when calculated with the default roughness value for conifers by 1.7 % and 4.1%, respectively, while there was no difference in modelled NO_y. The small differences in the dry deposition modelled for birch compared to conifer forest are due to the much higher contribution of the surface resistance, R_c, compared to the atmospheric resistances (R_a and R_b). For example, for birch, monthly R_c values for SO_2 ranged from 128 to 137 s m^{-1}, while the combined R_a and R_b was only 11 s m^{-1}.

Calculated total acid deposition (dry and wet) for birch was only 0.3% lower than for conifer forest, partly for the reasons above, but mainly because of the dominance of wet deposition, which remained the same for all land covers.

Modelled dry and wet deposition to the study grid square declined from 2002 to 2020 (Table 2), driven by the projected reduction in UK emissions of SO_2, NO_x and NH_3 (Defra, 2007), which are 64%, 45% and 19% lower, respectively, in 2020 than in 2002 (2004 baseline for NH_3). Reductions in modelled dry deposition were higher for SO_x and NO_y (58% and 38%, respectively) than for NH_x (11%), with the same pattern evident for wet deposition estimates. Modelled wet deposition of SO_x, NO_y and NH_x for 2020 was 54%, 40% and 13% lower, respectively, than in 2002, while total acidic pollutant deposition was 36% lower in 2020. Wet deposition dominated total deposition estimates for 100% birchwood cover in both 2002 and 2020, with dry deposition comprising for both years 10-11%, 12% and 17% of total SO_x, NO_y and NH_x deposition, respectively.

Table 2

Dry deposition estimates for the grid square were modified to reflect the actual (5.6%) and planned (29%) areas of birchwood in 2002 and 2020. Despite the reductions in UK pollutant emissions between 2002 and 2020, the expansion of birchwood was predicted to result in a threefold increase in dry deposition by 2020 (Table 2). However, this effect was more than compensated for by the modelled decline in wet deposition, which resulted in total SO_x, NO_y and NH_x deposition reducing by 2020 by 52%, 38% and 8%, respectively. The relative contribution of dry deposition of SO_x, NO_y and NH_x to total deposition increased from 0.7%, 0.7% and 1.1% in 2002 to 3.0%, 3.8% and 5.6% in 2020, respectively.

3.2. Streamwater chemistry and critical load exceedance

Mean high flow pH in the study catchments ranged from 5.6 to 6.4, with pH below 5.5 only measured once in a sample from LK2 (Table 3). The most strongly acidified stream appeared to be LK2, with a mean high flow alkalinity of -11.0 μeq l^{-1}, probably due to the higher proportion in this catchment of poorly-developed peaty ranker soils with a low mineral content and buffering capacity for acidity (see Table 1). Alkalinity and ANC determined in streamwater varied between high flow sampling occasions, most likely because the 9-10 sampling occasions were spread out over six separate months and winter, spring and autumn seasons. Mean streamwater ANC values were all positive, ranging from 36.1 to 74.8 μeq l^{-1}, and above the ANC_{crit} value of 0 μeq l^{-1} selected for this study which provides 90% or greater protection for brown trout at high flows. However negative ANC values were recorded in three of the study catchments on some occasions, suggesting that the fish population remained at risk from acid episodes. This was in line with the results of streamwater macroinvertebrate surveys (Gagkas et al., 2010). Streamwater marine SO_4, non-marine SO_4 (xSO_4) and NO_3 concentrations were relatively low, with marine SO_4 concentrations exceeding xSO_4 concentrations, reflecting a maritime dominance of atmospheric deposition (the nearest coast is 57 km to the west). The DOC concentrations measured in high flow samples on one occasion were moderate and similar in all catchments, ranging from 7.99 mg l^{-1} in LK5 to 9.01 mg l^{-1} in LK1.

Table 3

Critical loads calculated for each stream based on the mean high flow chemistry ranged from 1.51 (LK2) to 2.59 (LK5) keq ha^{-1} $year^{-1}$ of H^+ (Fig. 2). These were compared with the modelled total deposition of SO_x, NO_y and NH_x (Table 2) to determine the extent of CL exceedance in 2002 and 2020 (Fig. 2), and the impact of the projected expansion in birchwood cover. Critical loads were not exceeded in any of the study catchments in either of the modelled years, with the margin of non-exceedance increasing from 2002 to 2020, from 21% in LK5 to 55% in LK2 (Fig. 2). Therefore, the level of woodland expansion being planned (approximately a further 20%) was evaluated not likely to pose a risk of increased acidification in the study catchments if the reductions in acidic emissions projected for 2020 are realised.

Figure 2

4. Discussion

4.1. Influence of birch roughness length on modelled dry deposition

Although the roughness length (z_0) calculated in this study for birch and the default FRAME conifer value differed (0.73 vs. 1.0 m), they are more similar compared to those for shorter vegetation covers in FRAME, arable, grass and moorland, which have z_0 values of 0.03-0.05 (Singles, 1996). The relative similarity of the z_0 values for birch and conifer partly explains the very small difference in modelled dry pollutant deposition, but it is also due to the dominant influence of surface resistance (R_c) on the modelling of pollutant dry deposition compared to the contributions of the aerodynamic and laminar boundary layer resistances ($R_a + R_b$), particularly for N species. There are a number of uncertainties in the calculation of z_0 with the R94 model associated with the default constant values used, which can vary considerably between different vegetation types (Verhoef et al., 1997). Nevertheless, the limited influence of the calculated z_0 value on modelled dry deposition suggests that these uncertainties should not significantly affect the results of this study. However, to assess more accurately the effect of the planned expansion of broadleaf woodlands in the UK on pollutant dry deposition, operational atmospheric pollutant deposition models, such as FRAME, may also need to include a separate broadleaf land cover type with a suitable parameterisation of the surface resistance (R_c).

4.2. The effect of birchwood expansion on pollutant deposition

Modelled dry deposition onto the birch canopies contributed only a very small proportion of total deposition to the study grid square in both the 2002 and 2020 scenarios. This is partly because wet deposition is dominant, due to the area's high rainfall, and also because the study area is distant from the major pollution sources and areas of dry deposition of SO_2 and NO_y in the UK, i.e., the major urban and industrial areas in England and the interconnecting motorways. The pattern of wet SO_x deposition in the UK is less correlated to the sources, being a product of both local washout of the soluble gases SO_2 and H_2SO_4 and of the longer-range transport of secondary SO_4 aerosol (Dore et al., 2007). Due to its distance from pollutant sources the latter process is expected to dominate wet deposition of SO_x in the grid square studied. Similarly, wet deposition of NO_y in the UK does not occur close to sources but in areas with high precipitation, such as the uplands of Scotland and Wales (Dore et al., 2007). Dry deposition of NH_3 is more spatially variable because it is emitted close to the ground and is rapidly deposited onto vegetation, and consequently is highest close to sources, particularly intensive agricultural activities (Sutton et al., 1998). Since there are no such sources in the study area most NH_x deposition could be assumed to be associated with the long-range transport of NH_4 aerosol and wet deposition in upland areas with high annual precipitation (Dore et al., 2007).

Despite the FRAME model predicting that birchwood expansion would increase dry deposition by over three-fold, the dominance of wet deposition in the study area resulted in this having little effect on the modelled total deposition. Since the study area mean annual precipitation (2275 mm) is much higher than the UK average (c.1000 mm) the relative contribution of dry deposition and the significance of woodland expansion are expected to be greater in drier regions of the UK and also in areas closer to major pollutant sources. However, the enhancement of dry deposition of pollutants by woodland should continue to decline as a result of further planned reductions in emissions in the UK.

4.3. The effect of birchwood expansion on streamwater acidification

Recent (Matejko et al., 2009) and projected decreases in acidic emissions in the UK (Defra, 2007) should ensure that the study catchments at Loch Katrine remain protected from streamwater acidification, irrespective of the increased dry deposition that may occur due to the planned woodland expansion. This would remain the case even if the catchments were completely afforested with birch woodland. However, one area of uncertainty is the effect of birch woodland expansion on base cation cycling in the catchments. Increased base cation uptake by the new woodland (Reynolds, 2004) could further reduce the relatively low capacity of the study catchments for buffering acidic inputs, as indicated by the low streamwater alkalinity values and Ca concentrations at high flows. This will be offset by the intention not to harvest the native woodland, which will minimise base cation losses from the catchments. Furthermore, birch is thought to act as a soil improver (Miles, 1985) by increasing access through rooting to Ca in mineral subsoils and enhancing inputs to the surface soil by recycling via litterfall and canopy leaching (Reynolds, 2004). This action could help to increase the buffering of acidic inputs compared to the existing grassland cover. Weathering release of Ca in soil and soil available Ca concentrations have been shown to differ significantly between different tree species (e.g., Quideau et al., 1996; Washburn and Arthur, 2003), whilst Brandtberg et al. (2000) reported that soil available Ca concentrations were significantly higher in mixed birch-Norway spruce stands than in spruce-only stands.

Another uncertainty is if future N deposition exceeds woodland needs, leading to N saturation and increased NO_3 leaching. Despite large reductions in emissions of NO_x (50%) and NH_3 (18%) in the UK in the past 20 years, total deposition of N has changed little and was almost a factor of three higher than deposition of S in 2006 (RoTAP, in press). The relative importance of N deposition compared to S, and consequently of NO_3 as an acidifying anion, is expected to increase, although there is considerable uncertainty about the future behaviour of N in catchments (RoTAP, in press). Nitrate concentrations in streamwater during high flows have been shown to be significantly positively correlated with the percentage of mature broadleaf woodland cover in acid-sensitive catchments in the UK, with the highest concentrations occurring in catchments with >30% broadleaf woodland cover (Gagkas et al., 2008). Nitrogen uptake for tree growth will help to reduce the risk of N saturation, although N demand will decrease as the woodland matures, unless renewed through thinning and woodland regeneration.

In addition to the uncertainties identified above in the processes controlling the interaction between broadleaf woodland and streamwater acidification, there are also uncertainties relating to the impact of climate change on acid deposition and acidification (such as changes in hydrological regimes and in sea-salt deposition, Kernan et al. (2010)), and the representation of processes in critical load models. Improved representation of many terms, which are currently formulated rather simply, could be accommodated within the FAB model structure, for example, the influence of dissolved organic carbon (DOC) can be accommodated through reformulating the calculation of ANC (Lydersen et al., 2004). Other FAB model terms whose representation could be improved include: (1) N leaching (Kernan et al., 2010; Gagkas et al, 2010); (2) anion and base cation inputs in seasalt events (Cresser, 2007; Gagkas et al, 2010) which result in lower ANC values than expected; (3) DOC, which is widely increasing in concentration and may affect the acidity and recovery of freshwaters (Curtis et al., 2005).

4.4. Improving the modelling of dry deposition onto woodland canopies

The FRAME model used in this study to calculate dry pollutant deposition onto woodland canopies and CL exceedances in the study catchments contains several weaknesses for this application, which increases uncertainty in model predictions. One of these weaknesses is the use of fixed deposition velocities for particles, which means that the expected enhanced deposition of ammonium nitrate and ammonium sulphate particles as canopy roughness increases cannot be simulated in FRAME. Another possible weakness is the lack of explicit representation of the process of occult deposition, the scavenging of acidic pollutants in cloud water by vegetation canopies, which has been demonstrated to make a significant contribution to S and N inputs to upland conifer forests in the UK (Fowler et al., 1989). However, cloud water deposition is only important in the UK at altitudes >600 m (RoTAP, in press) where woodland expansion will be minimal and where any enhanced deposition effect may be counteracted by reduced tree growth and lower tree height. The model formulation also does not account for the altitude or spatial variability in the grid square of woodland cover (whether the woodland is present in one or more major blocks or as many small dispersed blocks with a larger forest edge length) and stand structure. Pollutant scavenging by woodland is expected to increase with altitude due to higher wind speeds and greater cloud cover. Dry deposition is known to be enhanced along woodland edges, especially for NH_3 in areas closer to intensive agricultural activities (Dragosits et al., 2002; RoTAP, in press), although this process is expected to be of minor importance in the study area where there are no major NH_3 sources. Variations in stand structure will affect the canopy roughness and hence the deposition velocity of pollutants, with greater deposition expected to woodland of mixed age and height, compared to more uniform woodland stands. However, to incorporate variability in woodland cover within grid squares would require a major restructuring of FRAME. Improving the accuracy of the acid deposition estimates used in CL exceedance calculations (such as the N_{dep} and S_{dep} terms in the FAB model) would help to reduce the uncertainty in assessing the risk of woodland expansion contributing to increased acidification, and increase confidence in sustainable forestry management and the protection of soil and water resources.

5. Conclusions

This study combined national scale methods, the FRAME atmospheric deposition model and the streamwater critical loads assessment procedure of the Forests & Water Guidelines, with a new calculation of the roughness length of birch, to assess whether broadleaf woodland expansion would have a significant effect on acid dry deposition and streamwater acidification at a specific site in Scotland. Despite their limitations, these methods were chosen because of their use in government policy applications in the UK and in similar forms in the UNECE. The main outcome of this study, that broadleaf woodland expansion had no significant effect on acid dry deposition and streamwater acidification in an area in which wet deposition is dominant, is relevant to forestry policy makers, managers and ecologists. The result will inform forestry best practice guidance documents and environmental statements for future planned woodland expansion schemes in similar climates, geologies and deposition regimes.

Acknowledgements

Z. Gagkas was funded by the Greek State Scholarships Foundation. The authors are grateful to the following for additional assistance and funding: The Forestry Commission; the British Atmospheric Data Centre (BADC); Ordnance Survey/EDINA; Dr R. Smith (Centre of Ecology and Hydrology, Edinburgh); Dr A. Lilly (The Macaulay Institute, Aberdeen); Dr B. Gardiner (Forest Research, Edinburgh); Mr. R. Lamont (Forest Enterprise, Aberfoyle); Mr. A. Gray, Mr.

J. Morman, Dr P. Anderson and Dr L. Eades of The University of Edinburgh; The University of Edinburgh Development Trust.

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Appendix

Calculation of birch roughness length from Raupach's simplified drag-partition model (R94, Raupach, 1994)

Roughness length, z_0, is related to the bulk drag coefficient, γ, of a rough surface at mean tree height, h (10.2 m in this study), and to the zero-plane displacement height, d:

$$\frac{z_0}{h} = \left(1 - \frac{d}{h}\right) \times e^{(-\kappa\gamma + \Psi_h)} \qquad \text{Eq. (A.1)}$$

which, when solved for z_0, becomes:

$$z_0 = (h - d) \times e^{(-\kappa\gamma + \Psi_h)} \qquad \text{Eq. (A.2)}$$

where κ (≈0.4) is the von Karman constant. Ψ_h, representing the influence of vegetation roughness on the velocity profile immediately above it, here took the value of 0.19, calculated in R94 with a standard formula using an empirical constant c_w. In R94 γ is approximated using the canopy area index, Λ, defined as the total (one-sided) area of all canopy elements per unit ground area:

$$\text{if } \Lambda > 0.6 \Rightarrow \gamma = \frac{1}{\sqrt{(C_S + C_R) \times 0.3}} \text{ else } \gamma = \frac{1}{\sqrt{(C_S + C_R) \times \frac{\Lambda}{2}}} \qquad \text{Eq. (A.3)}$$

For birch trees which are assumed to present the same frontal area in all directions, $\Lambda = 2\lambda$ and the empirically-derived constants C_S and C_R take values of 0.003 and 0.3, respectively. The roughness density or frontal area index, λ, is a key parameter of R94 and is defined by:

$$\lambda = \frac{1}{S}\sum_{i=1}^{n}\left(b_i \times S_t\right) \times c \qquad \text{Eq. (A.4)}$$

where S is the grid square area (25 km^2), c is an empirical coefficient that takes account of the cross-sectional shape of the tree crown and b_i is the area of vertical projection of the tree canopy within area S. Here $b_i = S$ because 100% cover was assumed in the calculations in FRAME. The dimensionless coefficient, S_t, which adjusts the frontal area to account for streamlining (Gardiner et al., 2000) was calculated from Eq. (A.5) which was parameterised from the results of wind tunnel studies conducted on paper birch (*Betula papyrifera*) (Vollsinger et al., 2005):

$$S_t = 2.47 \times U_h^{-0.83} \qquad \text{Eq. (A.5)}$$

where U_h, the mean wind speed at the top of the canopy, was taken as 10 m s^{-1} for the study area based on analysis of a 10-year dataset of 6-hourly windspeed data for the British Isles (Dore et al., 2006b).

Finally, in order to solve Eq. (A.2), the zero-plane displacement height, d, is calculated by:

$$d = \left(1 - \frac{1 - e^{-\sqrt{c_{d1}\Lambda}}}{\sqrt{c_{d1}\Lambda}}\right) \cdot h \qquad \text{Eq. (A.6)}$$

where c_{d1} is a fixed parameter with value 7.5.

Fig. 1. Location of the 5 km x 5 km grid square for which acid deposition was modelled and map of the boundaries of the study catchments showing the present day birchwood cover, estimated from aerial imagery, and the area of birchwood expansion in 2020 (see text for explanation).

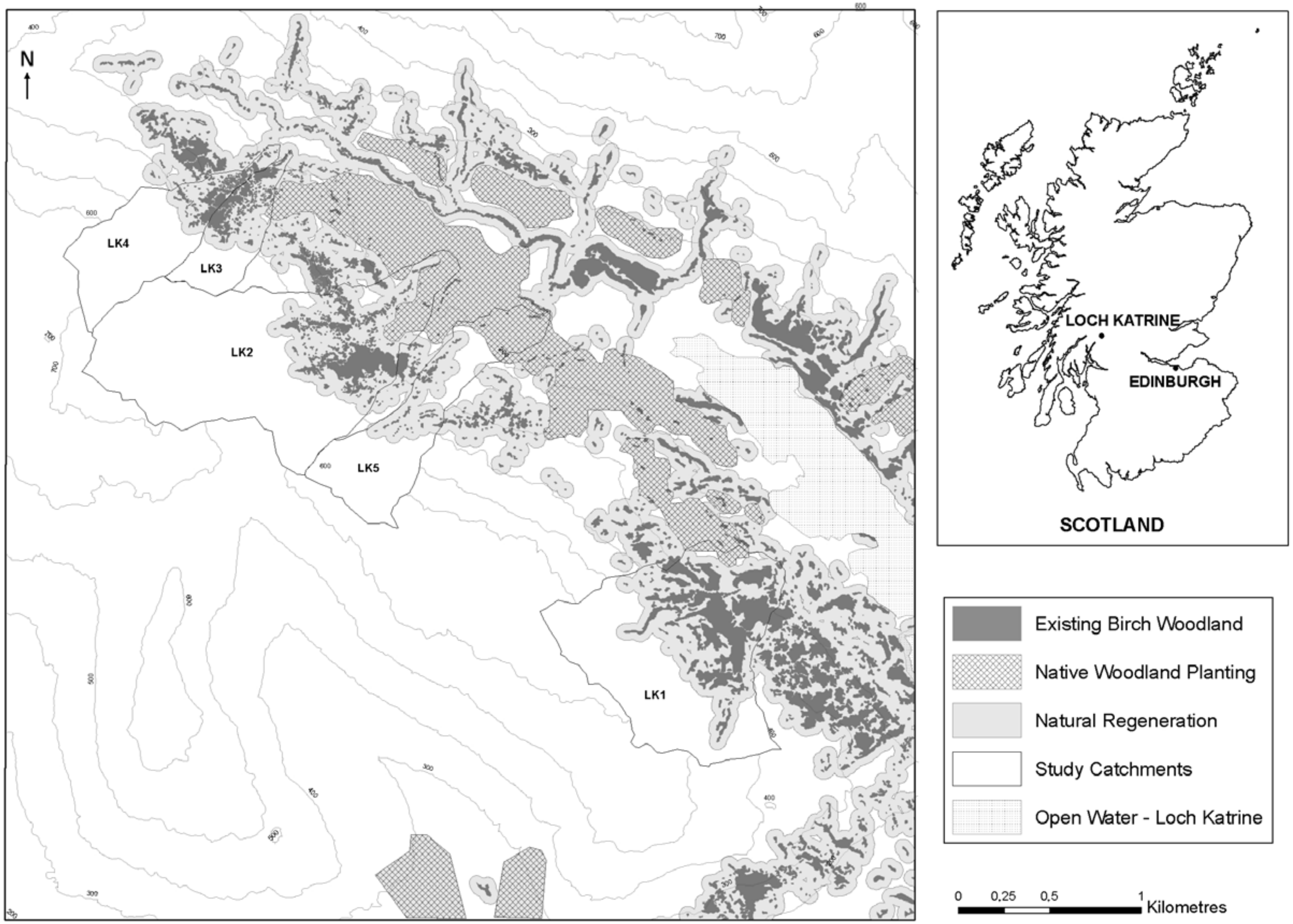

Fig. 2. CL and CL exceedance values calculated using the FAB model for the study catchments. Exceedances were calculated for 2002 and 2020 using the atmospheric deposition generated in the FRAME model from estimated emissions and birchwood cover for each of these years. Negative values of exceedances indicate non-exceedance. Catchment acronyms are given in Table 1.

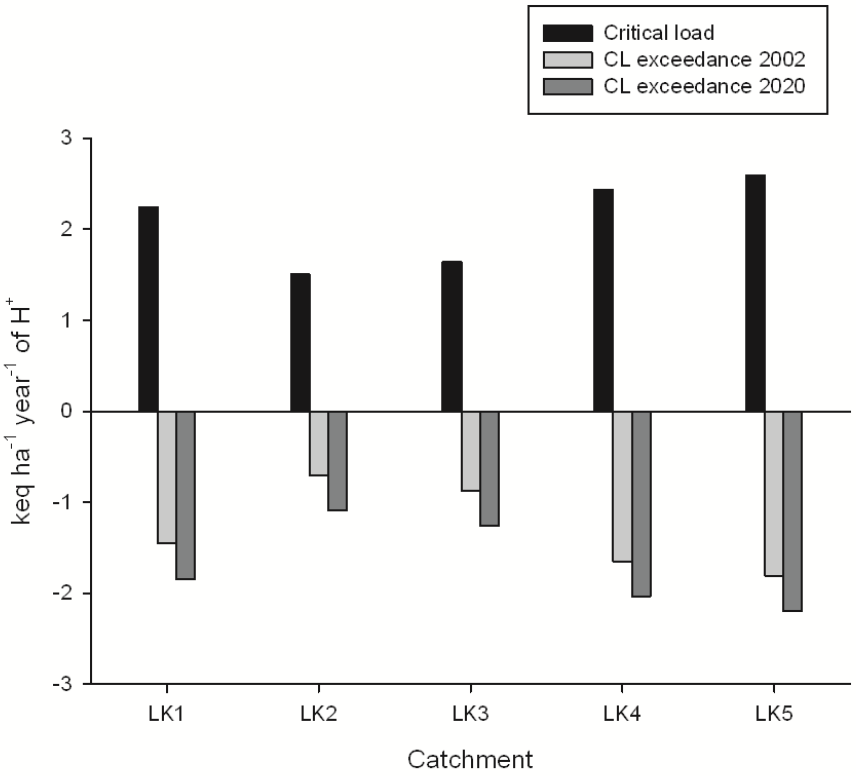

Table 1. Characteristics of the study catchments. Percentage birch woodland cover calculated from aerial imagery (Forest Enterprise, Aberfoyle, 1:10 000). Catchment areas, altitude and slope calculated from digital elevation models (Ordnance Survey/EDINA, Land-Form PROFILE®, 1:10 000). Catchment percentage cover of main soil types (PZ=podzols, PR=peaty ranker, GL=gleysols) from the National Soil Map for Scotland (MISR, 1981) (1:250 000).

Catchment	Present day birch woodland cover (% catchment area)	Catchment area (ha)	Mean (min-max) altitude (m)	Mean slope (º)	Cover of main soil types (%)
LK1	20.6	103	412 (128-683)	26	PZ (90), PR (8), GL (2)
LK2	10.4	132	461 (139-763)	23	PZ (81), PR (18), GL (1)
LK3	20.0	20.9	367 (185-556)	24	PZ (93), PR (0), GL (7)
LK4	8.14	39.6	502 (182-726)	26	PZ (88), PR (8), GL (4)
LK5	2.64	47.6	407 (134-681)	24	PZ (91), PR (4), GL (5)

Table 2. Dry, wet and total deposition of SO_x, NO_y and NH_x (in 10^{-3} keq ha^{-1} $year^{-1}$ of H^+) generated by the FRAME model for 2002 emissions and early 2000s birchwood cover and for 2020 emissions and projected birchwood cover in the study grid square. Figures for modelled dry deposition assuming 100% birchwood cover of the grid square are given in parentheses. Wet deposition estimates are unaffected by land use composition within the grid square.

Deposition type	2002 emissions & birch cover	2020 emissions & birch cover	% increase/ decrease
Dry deposition			
SO_x	3.1 (54)	7 (23)	+112 (-58)
NO_y	2.5 (45)	8 (28)	+222 (-38)
NH_x	3.0 (56)	14 (49)	+372 (-11)
Dry deposition total	*8.7 (155)*	*29 (100)*	*+235 (-35)*
Wet deposition			
SO_x	452	210	-54
NO_y	335	202	-40
NH_x	274	241	-13
Wet deposition total	*1060*	*654*	*-38*
Total deposition (dry + wet)	**1070**	**682**	**-36**
Dry deposition as % of total deposition	**0.8 (13)**	**4.2 (13)**	**---**

Table 3. Mean pH and concentrations of Gran alkalinity, Ca, Mg, Na, K, Cl, marine and non-marine sulphate (xSO$_4$), NO$_3$ and calculated ANC (all μeq l^{-1}) in streamwater sampled at winter high flow in the study catchments. Figures in parentheses are min and max values.

Catchment	No. samples	pH	Alkalinity	Ca	Mg	Na	K	Cl	SO$_4$	xSO$_4$	NO$_3$	ANC
LK1	10	6.2 (5.7-6.6)	41.6 (15.2-97.5)	77.2 (51.9-93.3)	41.8 (33.8-47.7)	117 (99.6-143)	8.03 (6.14-11.0)	135 (95.1-205)	40.9 (31.3-51.7)	27.0 (12.4-38.2)	8.80 (4.11-13.7)	60.8 (-12.4-87.0)
LK2	10	5.6 (5.2-5.8)	-11.0 (-45.3-10.0)	44.3 (29.4-55.5)	36.8 (29.2-41.1)	109 (96.1-134)	6.71 (4.60-10.2)	120 (90.0-185)	34.4 (30.1-42.6)	22.0 (11.0-31.5)	6.59 (<0.30-12.1)	36.1 (-25.4-55.9)
LK3	10	6.1 (5.9-6.5)	2.51 (-28.9-26.2)	54.3 (36.9-69.4)	37.0 (29.2-42.0)	120 (101-151)	6.94 (4.60-10.7)	134 (91.4-201)	38.7 (30.2-46.2)	24.8 (12.0-33.1)	5.39 (<0.30-12.2)	40.4 (-18.0-55.6)
LK4	10	6.3 (5.9-6.5)	36.7 (17.4-61.0)	78.8 (56.9-94.8)	44.7 (35.5-51.0)	113 (94.8-136)	6.83 (5.12-8.95)	125 (91.2-187)	41.4 (33.4-49.8)	28.4 (15.2-37.3)	11.4 (5.49-16.8)	65.7 (0.41-94.9)
LK5	9	6.4 (6.1-6.6)	31.7 (-12.2-72.8)	71.1 (50.4-82.8)	45.4 (36.2-49.4)	114 (97.4-138)	8.24 (6.14-12.3)	118 (93.2-149)	39.5 (34.0-45.2)	27.3 (18.7-33.8)	7.09 (2.16-13.4)	74.8 (54.6-92.8)